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(54) Title: UNIVERSAL COMPATIBILIZING AGENT FOR POLYOLEFINES AND POLAR PLASTICS

(57) Abstract: Product, method and use of a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D. The method of making AB comprise the steps of: a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group); together with maleic anhydride grafted polypropylene (B) allowing condensation reaction to occur between the functions under "A" and the anhydride under "B"; b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.



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Universal compatibilizing agents for polyolefines and polar plastics

Promoting the adhesion between two immiscible polymers has been a concern of increasing interest. Most useful applications are polymer alloying in very fine dispersion of one polymer into the other, and laminated structures and profiles obtained by coextrusion or colamination for example.

Various solutions have been proposed to induce some degree of interfacial bonding between two polymers exhibiting no or poor mutual adhesion. Among others, a favorite route is interleaving a third polymeric component whose segments provide good spontaneous adhesion with either component respectively. Master examples are di-block copolymers made by anionic polyaddition [references: S. H. Anastasiadis, I. Gancarz and J. T. Koberstein, *Macromolecules* **1989**, 22, 1449-1453; C. Creton, E. J. Kramer, C.-Y. Hui and H. R. Brown, *Macromolecules* **1992**, 25, 3075-3088]. Unfortunately, none of the existing commercial block copolymers is suitable to provide enough bonding strength between a polyolefine D such as polypropylene and a polar polymer C such as polyvinyl halide, polyvinylidene halide, poly(meth)acrylate, polyester, or polycarbonate.

It has been found surprisingly that a suitable compatibilizing agent AB of binary or ternary mixtures comprising at least one compound of the two above cited series of polymers C and D, can be obtained by the following way :

i. A copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein the functional moiety is characterized as being a hydroxyl, a carboxylic, a glycidyl or an amine functional group.

ii. The copolymer A described under "i" is melt processed together with maleic anhydride grafted polypropylene (B) allowing condensation reaction to occur between the functions under "i" and the anhydride under "ii".

iii. The new segmented multiblock copolymer obtained under "ii", is further melt processed together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.

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Accordingly, the present invention provides a method of making a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D comprising the steps of:

- 10 a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group);
- together with maleic anhydride grafted polypropylene (B) allowing condensation reaction to occur between the functions under "A" and the anhydride under "B";
- 15 b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.

20 In a preferred embodiment of the invention, the copolymer A contains 0.02 to 6 (x) mole percent of structural units bearing the functional group, the grafted copolymer B contains from 0.2 to 3 (y) mole percent of maleic anhydride units, the ratio of copolymer A and grafted copolymer B in the multiblock copolymer AB is such that the ratio x over y lies between 10 and 1 and preferably between 6 and 2.

25 In another preferred embodiment of the invention, the copolymer A contains between 0.1 to 6 mole percent of 2-hydroxyethyl(meth)acrylate and its weight average molecular weight (measured as PS equivalent molecular weight) is between 40000 and 500000 g/mol, preferably between 150000 and 500000 g/mol.

30 The copolymer A is easily obtained for example by solution polymerization in a suitable solvent such as methylethylketone (MEK), using a thermal initiator.

The present invention also provides a use of copolymer AB as defined above as a compatibilizing agent between a polar thermoplastic polymer C like polyvinyl halide homo or copolymer, polyvinylidene halide homo or copolymer, poly(meth)acrylate, polyester, or polycarbonate and a polyolefine D as single polymer or in blend with other polyolefines.

Polyolefine D can be polypropylene homo or copolymer with one of more co-monomer and made either with Ziegler-Natta catalyzer or with single site catalyzer. Polypropylene copolymer can have random or multisequence structure including the reactor made copolymer known as block copolymer.

Polyolefine D can also be a homo polyethylene or a co-polyethylene with zero to 50 wt % of a co-monomer with a number of carbon atoms from 3 to 20.

Endly polyolefine D can be a blend of a polymer D defined above with a polyolefinic rubber like EPDM, butyl rubber, BR, SBR.

Maleic anhydride grafted polypropylene is available in various grades and can be supplied by DuPont under the trademark FUSABOND®, by Exxon under the trademark EXXELOR®, by Uniroyal under the trademark POLYBOND®, by Atofina under the trademark OREVAC™ for example.

Example 1:

In a 2 liter stirred reactor, 840 g of methylethylketone, 274 g of methylmethacrylate, 6 g of 2-hydroxyethylmethacrylate (HEMA) and 2.8 g of azobis(isobutyronitrile) are introduced. The oxygen is removed by flushing dry nitrogen in the solution. The temperature is raised to 66 °C. The solution is reacted over 24 hours at 66 °C. An additional 1.4 g of azobis(isobutyronitrile) is added to the solution and the reaction is carried on during 12 more hours. The

copolymer is recovered by precipitation of the solution in deionized water as non solvent and then dried. The yield obtained is 95 % for the methylmethacrylate and 100 % for the HEMA. The copolymer is characterized
5 by weight average molecular weight of 152000 g/mol as measured by gel permeation chromatography.

The reactive melt processing leading to the copolymer AB is carried on by mixing 200 g of copolymer A, as prepared following the above description, and
10 100 g of copolymer B, FUSABOND® MD-353D, on a two roll mill at 180 °C during 30 minutes.

Sandwich elements consisting of a polypropylene sheet (1 mm thick) and a polyvinylidene fluoride sheet (2 mm thick) are bonded with a thin film (100
15 microns thick) of copolymer AB. The sandwiches are bonded by pressing the above construction during 15 minutes at 150 °C. The adhesion between the two sheets is tested by means of a U-peel test (reference: S. Wu, *Polymer interface and adhesion*, M. Dekker Inc., New York, 1982). A fracture energy (G) of 1607 J/m² has been measured.

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Examples 2 to 5:

In the example 2 to 4, the same experimental procedure as described in the example 1 has been followed. The HEMA amount introduced in the copolymer A is varied (see the following table) as well as the weight average molecular
25 weight, by means of a transfer agent (dodecanethiol-DDT). The measured fracture energies (G) are given for each example.

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Example number	mol % HEMA in		copolymer A		wt % DDT ^a	Mw
(g/mol) of copolymer A	x/y		G (J/m ²)			
2	1.68	0.49	94000	2.3	1036	
3	1.76	0.17	222000	2.4	1513	
5	4	5.09	0	172000	5.4	1659
5:without copolymer -					-	63

^a: This percentage is relative to the initial total mass of monomers used in the synthesis of copolymer A.

10 Example 6:

The blending of polyvinylidene fluoride with polypropylene is evaluated with and without using the copolymer AB. In particular, the blend of 19 weight percent of polyvinylidene fluoride, 76 weight percent of polypropylene and 5 weight percent of a copolymer AB (copolymer number 5 in the table presented above) prepared on a two roll mill (roll diameter: 110 mm; temperature: 180 °C; roll speed: 15 rpm; gap: 1 mm) leads to a two phase material presenting particles whose diameter ranges between 3 and 10 μm whereas the blending of polyvinylidene fluoride and polypropylene without copolymer displays particles characterized by a larger diameter, between 15 and 80 μm .

CLAIMS

1. Method of making a copolymer AB as a compatibilizing agent between a polar thermoplastic polymer C and a polyolefine D comprising the steps of:
- 5 a) melting a copolymer (A) comprising a larger amount of methylmethacrylate units and a smaller amount of functional vinyl or (meth)acrylate units wherein functional moiety is selected in the group consisting of (a hydroxyl, a carboxylic, a glycidyl or an amine functional group);
- together with maleic anhydride grafted polypropylene (B) allowing condensation
- 10 reaction to occur between the functions under "A" and the anhydride under "B";
- b) the condensation copolymer obtained under a) is further melt together with the two polymers C and D to be homogenized, or melt processed as an interlayer between the two molten polymers.
- 15
2. Method according to claim 1 wherein the copolymer (A) contains 0.02 to 6 (x) mole percent of structural units bearing the functional group, the grafted copolymer (B) contains from 0.2 to 3 (y) mole percent of maleic anhydride units, the ratio of copolymer A and grafted copolymer B in the multiblock copolymer
- 20 AB is such that the ratio x over y lies between 10 and 1 and preferably between 6 and 2.
3. Method according to claim 2 wherein the copolymer (A) contains between 0.1 to 6 mole percent of 2-hydroxyethyl(meth)acrylate and its weight average
- 25 molecular weight is between 40000 and 500000 g/mol, preferably between 150000 and 500000 g/mol.
4. Use of copolymer AB obtained by the method according to claim 1 to 3 as a compatibilizing agent between a polar thermoplastic polymer C and a
- 30 polyolefine D .

5. Use of copolymer AB according to claim 4 wherein the Polyolefine D is a polypropylene homo or a copolymer with one of more co-monomer.
- 5 6. Use of copolymer AB according to claim 4 wherein the Polyolefine D is a homo polyethylene or a co-polyethylene with zero to 50 wt % a co-monomer with a number of carbons from 3 to 20.
- 10 7. Use of copolymer AB according to anyone of claim 4 to 6 wherein the Polyolefine D is a blend with a polyolefinic rubber.
8. Use of copolymer AB according to Claim 7 with a rubber phase partially or fully crosslinked.
- 15 9. Use of copolymer AB according to Claims 4 to 8 with additive like inorganic filler and well known polymer stabilizers.
10. Copolymer AB obtained by the method according to anyone of claim 1 to 3

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B. FIELDS SEARCHED

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal, COMPENDEX

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	DATABASE WPI Section Ch, Week 199405 Derwent Publications Ltd., London, GB; Class A17, AN 1994-040099 XP002190638 & JP 05 345868 A (KURARAY CO LTD), 27 December 1993 (1993-12-27)	1,10
Y	abstract	1-10
Y	--- "MODIFICATION OF POLYAMIDE BY A CRYSTALLINE POLYOLEFIN AND GRAFT COPOLYMER" RESEARCH DISCLOSURE, KENNETH MASON PUBLICATIONS, HAMPSHIRE, GB, no. 332, 1 December 1991 (1991-12-01), pages 913-917, XP000274621 ISSN: 0374-4353 the whole document --- -/--	1-10

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>DATABASE WPI Section Ch, Week 198601 Derwent Publications Ltd., London, GB; Class A17, AN 1986-004879 XP002190639 & JP 60 233131 A (MITSUI TOATSU CHEM INC), 19 November 1985 (1985-11-19) abstract</p> <p>---</p>	1, 10
A	<p>"POLYPROPYLENE/POLAR POLYMER BLENDS" RESEARCH DISCLOSURE, KENNETH MASON PUBLICATIONS, HAMPSHIRE, GB, no. 330, 1 October 1991 (1991-10-01), pages 726-734, XP000264983 ISSN: 0374-4353 the whole document</p> <p>---</p>	1
A	<p>EP 0 335 649 A (ROHM & HAAS) 4 October 1989 (1989-10-04) claim 1</p> <p>---</p>	1
A	<p>DATABASE WPI Section Ch, Week 199051 Derwent Publications Ltd., London, GB; Class A17, AN 1990-380881 XP002190640 & JP 02 276841 A (OJI YUKA GOSEISHI KK), 13 November 1990 (1990-11-13) abstract</p> <p>---</p>	1
A	<p>DATABASE WPI Section Ch, Week 199211 Derwent Publications Ltd., London, GB; Class A14, AN 1992-085310 XP002190641 & JP 04 028743 A (SUMITOMO CHEM CO LTD), 31 January 1992 (1992-01-31) abstract</p> <p>-----</p>	1

INTERNATIONAL SEARCH REPORT

Information on patent family members

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Patent document cited in search report		Publication date	Patent family member(s)	Publication date
JP 5345868	A	27-12-1993	NONE	
JP 60233131	A	19-11-1985	NONE	
EP 0335649	A	04-10-1989	DK 552689 A	06-11-1989
			US 4957974 A	18-09-1990
			AT 141933 T	15-09-1996
			AU 3171689 A	05-10-1989
			BE 1003120 A5	03-12-1991
			CN 1041765 A	02-05-1990
			CN 1092782 A	28-09-1994
			DE 3910062 A1	26-10-1989
			DE 68927018 D1	02-10-1996
			DE 68927018 T2	20-03-1997
			DK 149389 A	30-09-1989
			EP 0335649 A2	04-10-1989
			FI 891479 A	30-09-1989
			FR 2629462 A1	06-10-1989
			GB 2216892 A , B	18-10-1989
			JP 2261811 A	24-10-1990
			JP 2022316 A	25-01-1990
			JP 2781197 B2	30-07-1998
			NL 8900757 A	16-10-1989
			NO 175537 B	18-07-1994
			NO 941044 A	27-12-1989
			US 5433984 A	18-07-1995
			US 5128410 A	07-07-1992
			US 5229456 A	20-07-1993
			US 5202380 A	13-04-1993
			AU 625059 B2	02-07-1992
			BR 8901431 A	14-11-1989
			IL 89774 A	13-05-1993
			IT 1232713 B	04-03-1992
			KR 179357 B1	15-05-1999
			NZ 228468 A	27-08-1991
			ZA 8902168 A	26-02-1992
			US 4997884 A	05-03-1991
			US 5035933 A	30-07-1991
			US 5147932 A	15-09-1992
			US 5300570 A	05-04-1994
JP 2276841	A		NONE	
JP 4028743	A	31-01-1992	JP 2897344 B2	31-05-1999